Transport imaging for contact-free measurements of minority carrier diffusion in GaN, GaN/AlGaN, and GaN/InGaN core-shell nanowires

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Minority carrier diffusion lengths (L_d) are measured for GaN, GaN/AlGaN, and GaN/InGaN core-shell nanowires using a technique based on imaging of recombination luminescence. The effect of shell material on transport properties is measured. An AlGaN shell produces L_d values in excess of 1 μm and a relative insensitivity to wire diameter. An InGaN shell reduces effective diffusion length, while a dependence of L_d on diameter is observed for uncoated nanowires. © *2011 American Institute of Physics*. [doi:10.1063/1.3573832]

Gallium nitride (GaN) nanowires have shown promise for applications in optoelectronics, high power, and high speed devices.[1–4] For many potential applications, the transport behavior of minority carriers affects device performance and serves as an indicator of material quality. Determination of minority carrier diffusion lengths (L_d) is generally performed using electron beam induced current (EBIC),[5–7] light induced transient grating,[8] or a combination of measurements of lifetime with estimates of minority carrier mobility.[9,10] Strengths and limitations of these approaches vary but most either require contacts or lack the spatial resolution for nanostructure characterization.

In this paper, we utilize an imaging approach that allows for a direct measure of L_d from an image of the luminescence associated with carrier diffusion and recombination. Near field scanning optical microscopy (NSOM) is used in conjunction with charge generation in a scanning electron microscope (SEM) to measure a series of core-shell nanowires and determine the effect of the shell material.

Samples are mounted in a Nanonics MultiView 2000 atomic force microscope (AFM). This instrument allows for independent scanning of both sample and fiber probe and direct electron beam access to the sample. In transport imaging, the NSOM probe is scanned while the incident beam is fixed at a point on the sample to generate excess carriers. The probes are cantilevered fibers with apertures ranging from 100 to 500 nm. Collection efficiency is strongly dependent on aperture size, so tradeoffs are required between AFM resolution and optical collection. For this work, we utilized tips of 200–300 nm diameter.

Transport imaging depends on maintaining the spatial distribution of the luminescence associated with carrier recombination. This is in contrast to standard cathodoluminescence (CL) in which the excitation point is scanned and all recombination light is mapped to the point of excitation. The fundamental approach is basically an optical Haynes–Shockley experiment.[11–13] While transport imaging in thin films can be done using far field techniques, the size of the structures and the small diffusion lengths observed in GaN nanowires require the resolution provided by near field imaging.[14,15]

We investigated GaN nanowires, as well as core-shell structures with GaN cores and either AlGaN or InGaN shells. GaN nanowires were grown by metal-catalyzed metal organic chemical vapor deposition on *r*-plane sapphire substrates coated with 2 nm Ni at a temperature of 900 °C. The resulting nanowires are single crystalline and have triangular cross sections with a $[11\bar{2}0]$ growth orientation and one $(000\bar{1})$ and two equivalent $\{1\bar{1}01\}$ facets.[3] The nanowires are unintentionally doped n-type with typical lengths of 5–30 μm and diameters of 100–800 nm. Further details on GaN nanowires grown by this method have been previously published.[16,17] Residual doping has been estimated to be $\sim 10^{17}$ cm^{-3} based on measured resistivity as a function of diameter and modeling of surface depletion effects.[17]

InGaN shell growth[18] was carried out at a temperature of 710 °C for 10 min. AlGaN shell growth was carried out at 1075 °C for 8 min. AlGaN layers ranged in thickness from ~10–25 nm with Al mole fraction ranging from 20%–30% as determined by cross-sectional scanning transmission microscopy and energy dispersive x-ray spectroscopy. The InGaN layers ranged in thickness from ~30–90 nm with In mole fraction of ~17%. Cross-sectional images of representative core-shell nanowires from the samples are presented in Ref. 19. Little to no InGaN growth is observed on the $(000\bar{1})$ facet,[18] whereas AlGaN growth is observed on all three nanowire facets.

Figure 1 shows 300 K CL spectra for the three samples: GaN/AlGaN, uncoated GaN and GaN/InGaN nanowires. The nanowires are dispersed on Si, so substrate luminescence is negligible. In all cases, the band edge luminescence associated with GaN is observed, along with broad defect-related luminescence. Yellow luminescence is present in all cases and has previously been shown to be predominantly from the surface layer in individual GaN nanowires, though also present in the center region.[20] No luminescence is evident from the shell layers.

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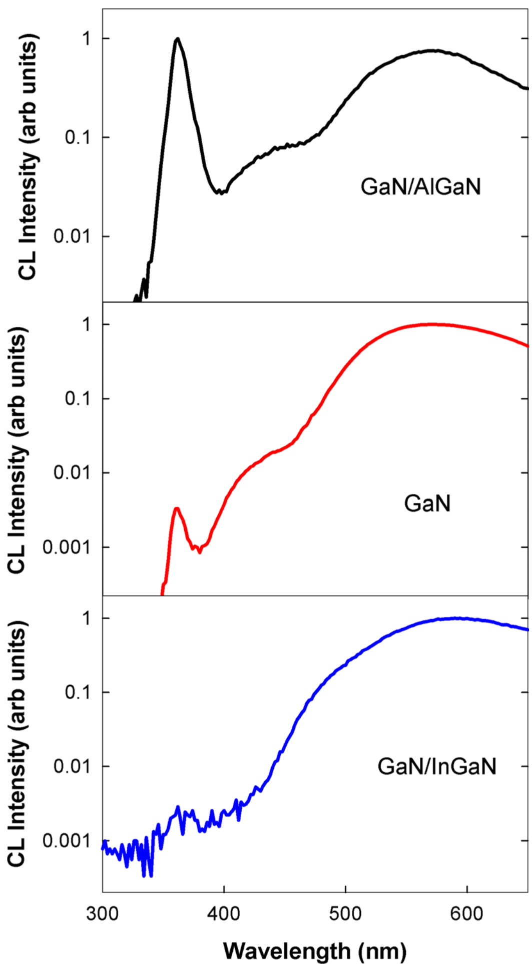

FIG. 1. (Color online) CL spectra of GaN/AlGaN, GaN, and GaN/InGaN nanowires. Spectra were taken at room temperature with electron beam energy of 20 keV. Note that the intensity is on a logarithmic scale.

For transport imaging experiments, a single nanowire is selected. The electron beam is incident at a fixed point. The NSOM tip is scanned and the panchromatic luminescence distribution is collected and displayed as intensity as a function of position.

Figure 2 shows the topography (left) and NSOM (right) distributions for an uncoated GaN wire. The point of carrier generation is just below the image and NSOM scanning is performed in the region adjacent to the incident electron beam. The wire diameter is ~500 nm, determined from the SEM image. The AFM image, while useful for determining the axis for extraction of the spatial variation in the intensity, is not a good indication of wire diameter due to the convolution of the NSOM probe (250 nm) with the nanowire.

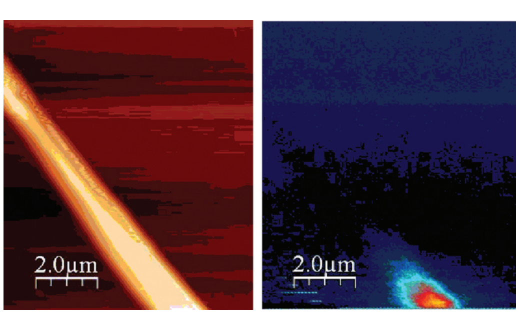

FIG. 2. (Color) Topography (left) and near field optical intensity (right) images during e-beam excitation of a GaN nanowire.

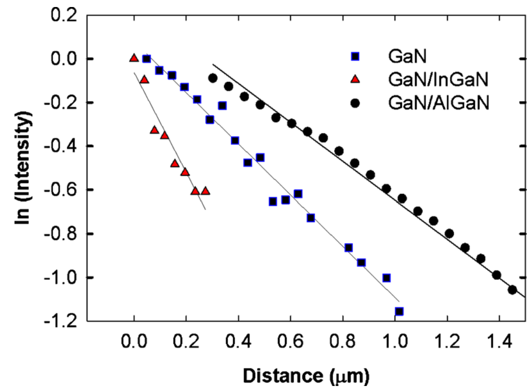

FIG. 3. (Color online) Luminescence intensity as a function of position for GaN/AlGaN (●), GaN (■), and GaN/InGaN (▲) nanowires. All nanowires are of diameter ~700±50 nm.

The highest luminescence intensity is observed, as expected, in the region adjacent to the point of excitation, decreasing with distance from that point. Figure 3 shows the intensity distributions as a function of position for GaN, GaN/AlGaN, and InGaN/GaN wires with ~700 nm diameter. Data are normalized to a starting intensity. The operation of the NSOM in the SEM limits the working distance and therefore the best resolution for nanowire diameter is ~50 nm.

For diffusion in one dimension, the minority carrier distribution and associated luminescence intensity distribution are given by $I(x)=(g/2L)\exp(-x/L_d)$ where I is intensity, g is generation rate, L is diffusion length, and x is the distance from the point of excitation. The diffusion length is obtained from the slope of a plot of ln(I) as a function of x. This yields an effective diffusion length, with the combined effect of recombination associated with both bulk and surface effects.

Measurements were made for the three types of wires described above. Table I summarizes the results. In all cases, the electron beam excitation energy was 20 keV and the probe current was 3×10^{-10} A. We use the lowest possible probe current in order to operate in the low excitation regime where the excitation does not significantly change the population of majority carriers. Consistent values of L_d for lower probe currents indicate that the low excitation limit has been achieved.

For nanowires of comparable diameter, the AlGaN shell produces the longest effective L_d, with the uncoated wires intermediate and InGaN/GaN showing the smallest value. This behavior is consistent with the role of

TABLE I. 300 K minority carrier diffusion lengths for GaN/AlGaN, GaN, and GaN/InGaN wires.

Wire	Diameter (nm)	Diffusion length (nm)
AlGaN/GaN-n-type	200	1100
	500	1200
	700	1100
GaN-n-type	300	130
	500	660
	700	830
InGaN/GaN-n-type	500	350
	700	430
	1000	470

AlGaN as a wider bandgap material, creating a barrier for recombination at the surface. InGaN, in contrast, has a smaller bandgap than GaN. Density functional calculations suggest a conduction band alignment of ~0.5 eV for a 17% In concentration.[21] The band bending resulting from this discontinuity at the InGaN/GaN interface will produce a depletion region in the wire that will enhance the diffusion of minority carrier holes to the surface. This is evident in both the decreased band to band CL and in the reduced effective L_d.

Within a given material system, we have imaged multiple wires to determine the effect of wire diameter. For the AlGaN/GaN wires, the dependence on wire diameter is small, again indicating the role of the AlGaN shell in reducing the effect of surface recombination. This is consistent with previous evidence for passivation of GaN nanowire surface states by an AlGaN shell.[22,23] For the uncoated GaN wires, however, L_d is reduced from 710 to 130 nm, as diameter decreases from 700 to 300 nm.

Since these are n-type wires, the diffusion lengths reflect the transport of holes. In MBE-grown structures, lifetimes ranging from ~0.5 to 3 ns were measured for Si doped wires with diameters ranging from 250 to 1000 nm.[24] Combining this with a reported value of hole mobility in GaN nanowires of 12 $cm^2/V\ s$,[25] one obtains an estimated L_d of 120 to 300 nm for an uncoated wire, consistent with the values reported here.

For comparison with transport imaging, we performed EBIC measurements on similar GaN/AlGaN and uncoated GaN wires. The contacts were Ni/Au with 40 nm of Ni deposited, followed by 70 nm of Au. Only one sample was available in each case. For the GaN/AlGaN nanowire, a plot of the collected current along the wire as a function of position[19] results in a characteristic decay length of 1.0 μm at the left side and 1.4 μm on the right side for a wire with diameter of ~800 nm. Since an ohmic contact is not available at one end of the structure, the magnitude of the EBIC current is affected by the contact behavior and values for diffusion length cannot be directly obtained.[26] However, the characteristic length determined from the decay of the collected current can provide an independent order of magnitude estimate for carrier diffusion. For the uncoated GaN wire, this results in a characteristic length of 260 nm for a ~200 nm diameter wire, significantly less than measured for the GaN/AlGaN structure of comparable diameter.

In summary, we have applied a near-field imaging technique to measure minority carrier diffusion lengths in GaN, GaN/AlGaN, and GaN/InGaN nanowires. This optical approach allows for direct measure of L_d without the need for contact fabrication. The results show that nanowires with an AlGaN shell have the longest diffusion lengths (~1.2 μm) for diameters ranging from 200 to 700 nm. For both GaN and GaN/InGaN structures, the effective diffusion lengths are reduced, most likely by surface recombination effects. Size dependent diffusion lengths are observed in the uncoated GaN structures, consistent with prior reports of diameter dependent lifetimes and mobilities in similar structures.

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[1]M. Law, J. Goldberger, and P. D. Yang, Annu. Rev. Mater. Res. **34**, 83 (2004).
[2]W. Lu and C. M. Lieber, J. Phys. D: Appl. Phys. **39**, R387 (2006).
[3]G. T. Wang, A. A. Talin, D. J. Werder, J. R. L. Creighton, E. Lai, R. J. Anderson, and I. Arslan, Nanotechnology **17**, 5773 (2006).
[4]S. Barth, F. Hernandez-Ramirez, J. D. Holmes, and A. Romano-Rodriguez, Prog. Mater. Sci. **55**, 563 (2010).
[5]L. Chernyak, A. Osinksy, H. Temkin, J. W. Yang, Q. Chen, and M. A. Khan, Appl. Phys. Lett. **69**, 2531 (1996).
[6]Z. Z. Bandić, P. M. Bridger, E. C. Piquette, and T. C. McGill, Appl. Phys. Lett. **72**, 3166 (1998).
[7]E. B. Yakimov, S. S. Borisov, and S. I. Zaitsev, Semiconductors **41**, 411 (2007).
[8]J. Mickevičius, M. S. Shur, R. S. Q. Fareed, J. P. Zhang, R. Gaska, and G. Tamulaitis, Appl. Phys. Lett. **87**, 241918 (2005).
[9]S. F. Chichibu, H. Marchand, M. S. Minsky, S. Keller, P. T. Fini, J. P. Ibbetson, S. B. Fleischer, J. S. Speck, J. E. Bowers, E. Hu, U. K. Mishra, S. P. DenBaars, T. Deguchi, T. Soto, and S. Nakamura, Appl. Phys. Lett. **74**, 1460 (1999).
[10]Y. Kawakami, K. Omae, A. Kaneta, K. Okamoto, T. Izumi, S. Saijou, K. Inoue, Y. Narukawa, T. Mukai, and S. Fujita, Phys. Status Solidi A **183**, 41 (2001).
[11]J. R. Haynes and W. Shockley, Phys. Rev. **81**, 835 (1951).
[12]D. R. Luber, F. M. Bradley, N. M. Haegel, M. C. Talmadge, M. P. Coleman, and T. D. Boone, Appl. Phys. Lett. **88**, 163509 (2006).
[13]N. M. Haegel, T. J. Mills, M. Talmadge, C. Scandrett, C. L. Frenzen, H. Yoon, C. M. Fetzer, and R. R. King, J. Appl. Phys. **105**, 023711 (2009).
[14]N. M. Haegel, C.-H. Low, L. Baird, and G.-H. Ang, Proc. SPIE **7378**, 73782B (2009).
[15]L. Baird, G. H. Ang, C. H. Low, N. M. Haegel, A. A. Talin, Q. Li, and G. T. Wang, Physica B **404**, 4933 (2009).
[16]Q. Li and G. T. Wang, Appl. Phys. Lett. **93**, 043119 (2008).
[17]A. A. Talin, G. T. Wang, E. Lai, and R. J. Anderson, Appl. Phys. Lett. **92**, 093105 (2008).
[18]Q. Li and G. T. Wang, Appl. Phys. Lett. **97**, 181107 (2010).
[19]See supplementary material at http://dx.doi.org/10.1063/1.3573832 for cross sectional STEM images of GaN/InGaN and GaN/AlGaN nanowires and EBIC of GaN/AlGaN.
[20]Q. M. Li and G. T. Wang, Nano Lett. **10**, 1554 (2010).
[21]P. G. Moses and C. G. Van de Walle, Appl. Phys. Lett. **96**, 021908 (2010).
[22]R. P. Prasankumar, P. C. Upadhya, Q. Li, N. Smith, S. G. Choi, A. K. Azad, D. Talbayev, G. T. Wang, A. J. Fisher, J. Hollingsworth, S. A. Trugman, S. T. Picraux, and A. J. Taylor, Proc. SPIE **7406**, 74060E (2009).
[23]A. Armstrong, Q. Li, Y. Lin, A. A. Talin, and G. T. Wang, Appl. Phys. Lett. **96**, 163106 (2010).
[24]J. B. Schlager, K. A. Bertness, P. T. Blanchard, L. H. Robins, A. Roshko, and N. Sanford, J. Appl. Phys. **103**, 124309 (2008).
[25]Z. H. Zhong, F. Qian, D. L. Wang, and C. M. Lieber, Nano Lett. **3**, 343 (2003).
[26]P. R. Wilshaw, Ultramicroscopy **31**, 177 (1989).